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A Summary of Radiation Damage Studies in Barium Fluoride from the GEM Collaboration

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A summary is given of the radiation damage studies in BaF_2 carried out by the GEM Collaboration.. Data are presented on the effects of radiation from low energy gamma rays, energetic neutrons and high energy hadrons. Results are given from various analytical techniques used to study crystal purity and structure, and the present understanding of the principle causes of damage is discussed. A brief summary is also given of the conclusions of an Expert Panel which reviewed the situation of radiation damage in BaF_2 for the GEM experiment.

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1. Introduction

Over the past several years, the GEM Collaboration has carried out an extensive study of radiation damage in BaF_2 as part of their program to develop a high resolution electromagnetic calorimeter for the SSC. This was motivated by the high radiation resistance requirements for the EM calorimeter in the strong radiation field produced at the SSC, along with the desire to achieve and maintain extremely good energy resolution over the lifetime of the experiment. Barium fluoride was chosen for this purpose because of its high intrinsic radiation resistance, and because of the short decay time of its fast component which allows the detector to operate at the very high rates expected at the SSC. However, because of the stringent demands on precision and stability, it was found that a a systematic study of the radiation effects in BaF_2 was required in order to meet the GEM requirements. This was undertaken by a team of scientists, engineers and crystal growers spanning many disciplines, and brought a tremendous amount of expertise to bear on a specific problem. The result was major improvement in radiation hardness of BaF_2, especially for large crystals, as well as a much better understanding of the principle causes of damage. Many of the results obtained in this study apply not only to BaF_2, but also to other crystals.

2. The GEM Detector

The GEM BaF_2 calorimeter is described in detail elsewhere in these proceedings [1].It consists of approximately 16,000 crystal pairs arranged in an array which covers an angular region of $\theta = \pm 175^o$ and $\Delta\phi = 2\pi$ located at a radius of 75 cm from the intersection region inside the GEM central detector. Each crystal pair consists of two 25 cm long trapezoidal crystals joined together at the center and read out with a UV sensitive photodevice at one end. The ionizing radiation dose for the calorimeter reaches levels of up to 3.5x10^6 rad/yr at the smallest angles, and $\sim 3x10^4$ rad/year for the central region. The fluence of neutrons reaches levels of up to $10^{14} n/cm^2$ at the smallest angles, and $\lesssim 10^{13} n/cm^2$ in the central region. These neutrons are produced mainly by hadronic showers in the hadron calorimeter located behind the BaF_2 detector and have energies typically around 1 MeV.

3. Radiation Damage from Gammas, Neutrons and Hadrons

A number of early studies of radiation damage in BaF_2 indicated that its intrinsic radiation hardness was quite good [2],However, most of these studies were done with small crystals and were not always carried out under controlled conditions. More recent measurements have shown that larger samples are much more susceptible to radiation damage, and that virtually all crystals exhibit damage to some degree [3-5].This damage generally appears as the production of absorption bands in the UV or visible part of the spectrum which affects the transmission of scintillation light through the crystal. So far, no evidence has been found that would indicate that the actual scintillation efficiency is affected by radiation, and, at least in large crystals, the apparent loss in scintillation light output can be totally explained by the decrease in transmission [4].

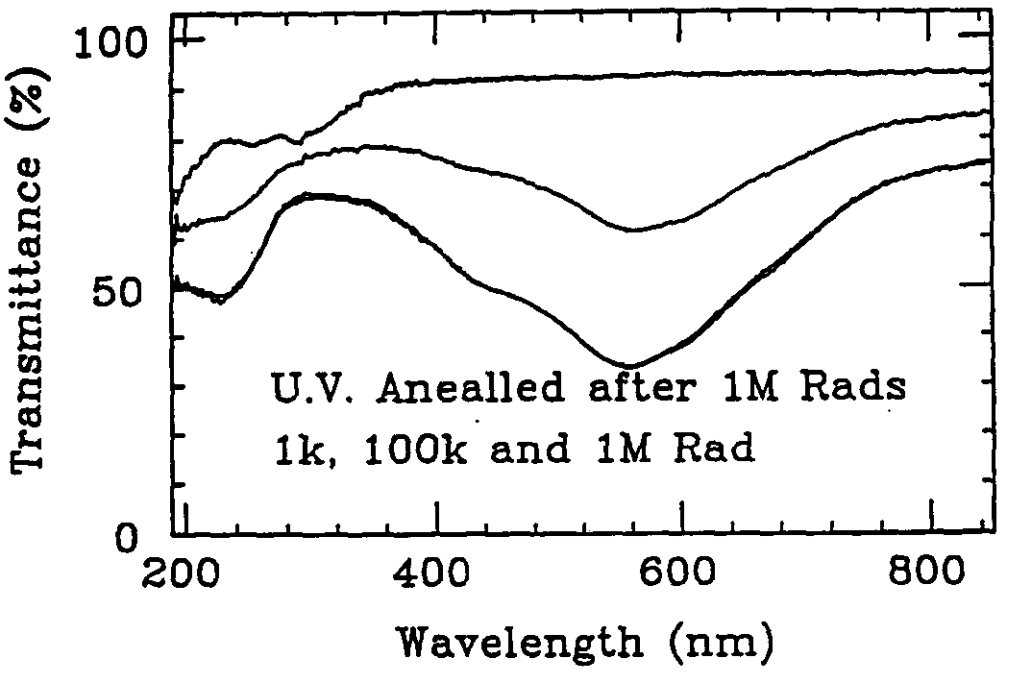

Fig.1 *Transmittance of a 25 cm long BaF_2 crystal (SIC 402) recently produced at the Shanghai Institute of Ceramics for various doses of ^{60}Co irradiation. The light attenuation length was ~ 41 cm at 220 nm after 1 Mrad. This crystal had also been previously irradiated to 1 Mrad and annealed with UV light.*

Figure 1 shows one of the most recent 25 cm long samples produced by the Shanghai Institute of Ceramics (SIC) which was irradiated to 1 Mrad with ^{60}Co gamma rays. A strong absorption band is produced at $\sim$ 560 nm, along with some additional induced absorption in the 200-220 nm range. In spite of the amount of damage produced in this crystal, it represents a substantial improvement in radiation hardness compared to many earlier long crystals, and comes very close to meeting the GEM specification of $\geq$ 70% transmission at 220 nm after 1 Mrad. As can be seen from the curves, the increase in absorption with dose saturates at a level of 100 Krad. This saturation effect is generally observed in all gamma ray irradiations and is characteristic of the kind of effect one would expect if the damage is being caused by a finite number of impurities or defects in the crystal. Another important feature of the damage produced by gamma rays is that it is fully recoverable. This recovery can be induced by thermal annealing, typically done by heating the crystal to $\sim 400^oC$ for several hours in dry nitrogen, or by optical annealing. Techniques for optical annealing will be further discussed below.

Radiation damage caused by neutrons has also been studied in the GEM BaF_2 crystals. Irradiations were carried out at University of Lowell Van de Graaff using neutrons with energies of a few MeV produced in the reaction $^7Li(p,n)^7Be$. The neutrons were accompanied by a flux of 429 keV gamma rays which were also produced in the reaction. The advantage of this type of neutron irradiation over those done at a reactor is the more energetic spectrum of neutrons, which more closely resembles the spectrum expected at the SSC, and the relatively low accompanying dose of gamma rays. The curve labeled n in Figure 2 shows the absorption induced in a 25 cm long crystal irradiated to a total fluence of $\sim 10^{13} n/cm^2$ with an accompanying gamma ray flux of $\lesssim$ 1 Krad. The other curves show the crystal after it was thermally annealed after the neutron irradiation and re-irradiated up to 1 Mrad with ^{60}Co gamma rays. The conclusions from these data are that the neutrons produce little or no additional damage beyond that which is caused by the accompanying gamma rays, and that the combined neutron plus photon damage is fully annealable.

Radiation damage caused by high energy hadrons was also studied in an measurement carried out at Brookhaven National Laboratory. Several BaF_2 crystals were placed inside one of the primary target caves of the Alternating Gradient Synchrotron and exposed to

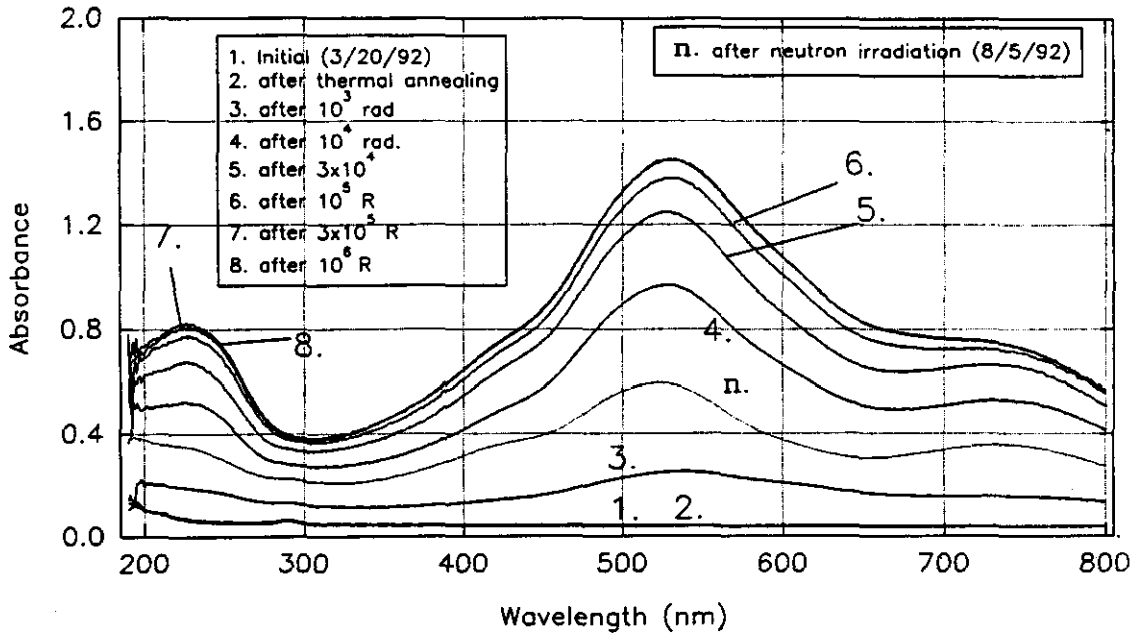

Fig.2 ***Absorbance vs. wavelength for a 25 cm long BaF_2 crystal (S3B009) irradiated with $\sim 10^{13} n/cm^2$ plus $\lesssim$ 1 Krad of gamma rays (curve labeled n). The crystal was then thermally annealed and then re-irradiated up to 1 Mrad with ^{60}Co gamma rays.***

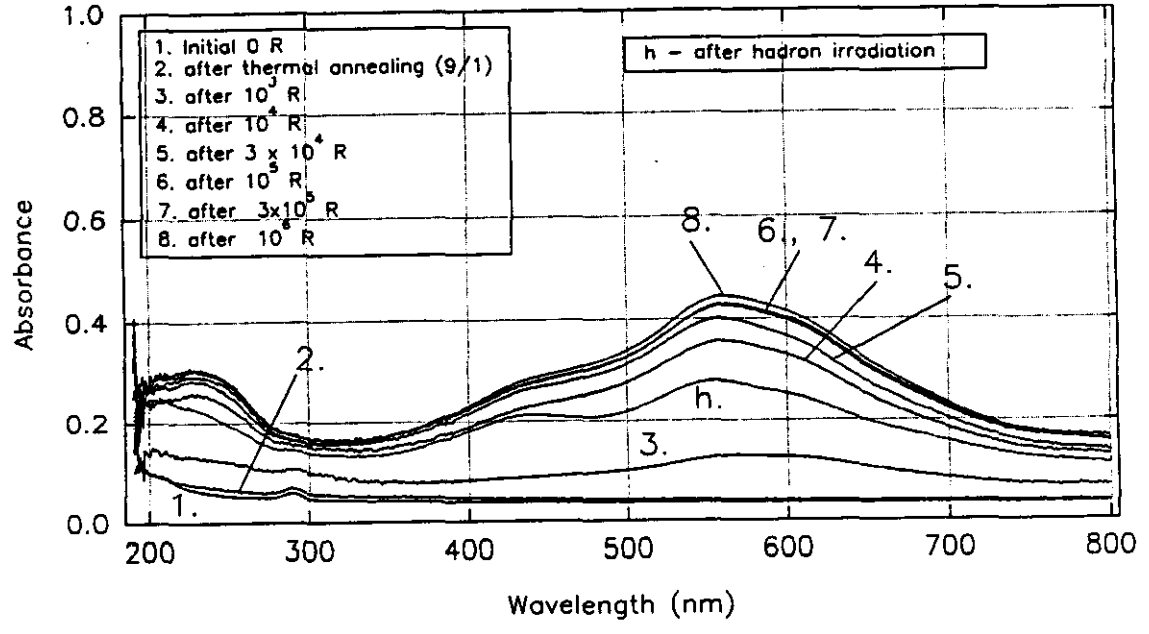

Fig.3 ***Absorbance vs. wavelength for a 20 cm long BaF_2 crystal (SIC 703) irradiated up to $\sim$ 2 Mrad with hadrons (curve labeled h). The crystal was then thermally annealed and then re-irradiated up to 1 Mrad with ^{60}Co gamma rays.***

showers of hadrons produced by 24 GeV proton-nucleus collisions. Figure 3 shows a 20 cm long crystal which received a total hadronic dose of $\sim$ 2 Mrad, indicated by the curve labeled *h*. The total dose, however, is uncertain by up to a factor of two because the dose was not measured in the BaF_2 directly, but rather using LiF dosimeters placed next to the crystals during the irradiation. The crystal was then thermally annealed and re-irradiated up to 1 Mrad with ^{60}Co gamma rays, as shown by the other curves in the figure. The conclusions from this study were that the damage caused by hadrons is less than or equal to the equivalent dose of low energy gamma rays, and that the hadron damage is also fully recoverable.

As stated above, it was found that recovery from damage could be quickly and effectively accomplished by thermal annealing at an elevated temperature. However, recovery is extremely slow at room temperature, especially in the short wavelength region. This is shown in Figure 4, which shows the change in transmission of a 25 cm long crystal initially irradiated to a dose of 1 Mrad and kept in the dark at room temperature for 260 days. There is a noticeable recovery in transmission at longer wavelengths, but only a small, yet discernable change in transmission at shorter wavelengths. However, recovery can also be induced by UV or visible light. Figure 5 shows the recovery in transmission as function exposure time to 400 nm light after irradiating a crystal to 1 Mrad with gamma rays. The transmission at 220 nm increases to a plateau after approximately 6 hours, at which point the attenuation length has reached a value of $\sim$ 180 cm, which is within the GEM specifications. This feature of recovery provides an important way of dealing with radiation damage in the actual experiment by allowing for the possibility of annealing the crystals *in situ* while the experiment is running. The fact that the optical annealing can be done with visible light implies that the readout device used for the crystals, which is not sensitive to visible light, would be blind to the light providing the annealing. This strategy was adopted by the GEM collaboration such that the calorimeter could be built with presently available crystals, such as the one shown in Fig. 1. It was also shown that the optical annealing for the entire electromagnetic calorimeter could be provided with only a few hundred watts of power [6].

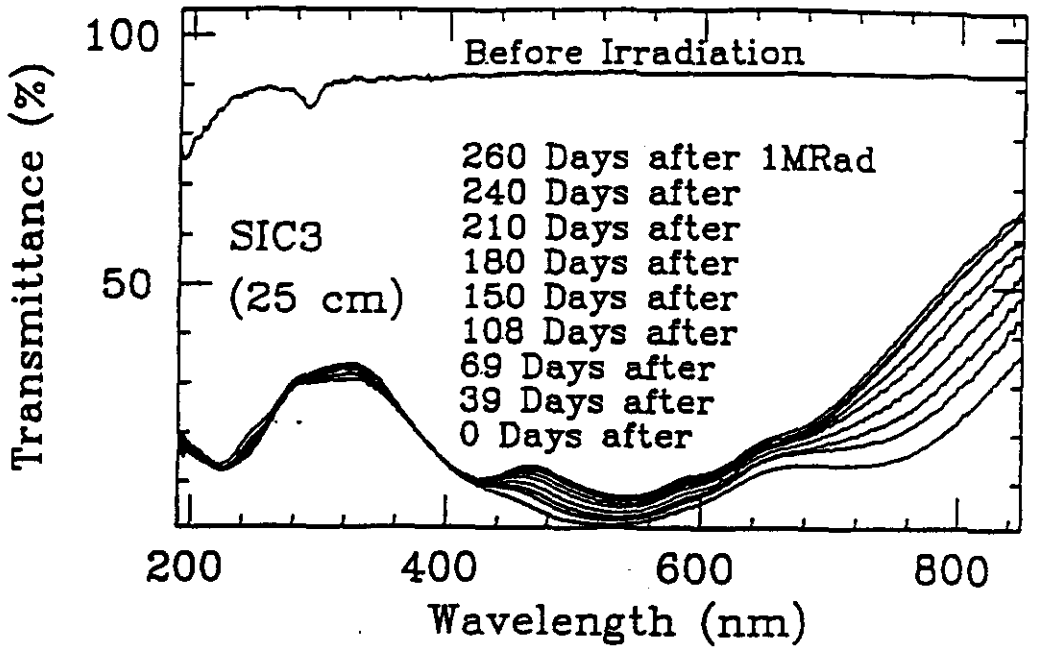

Fig.4 *Transmittance vs. wavelength for a 25 cm crystal (SIC3) irradiated to 1 Mrad with ^{60}Co gamma rays and kept in the dark for 260 days.*

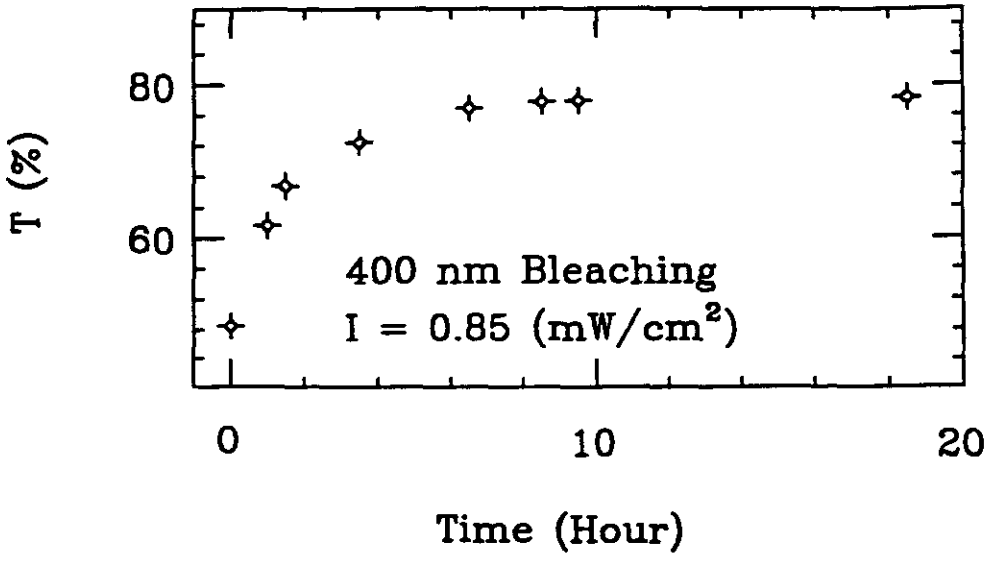

Fig.5 *Recovery in transmittance in a 25 cm crystal (SIC 402) after irradiation to 1 Mrad with ^{60}Co gamma rays with exposure to 400 nm light.*

A great deal more information on radiation damage was obtained during the course of the GEM study, and a detailed summary of recent results can be found in [7].Some of the main experimental facts are summarized below:

- Saturation is observed in transmission, light output, phosphorescence and thermoluminescence for irradiation with gamma rays. Saturation has not been established for neutrons and charged hadrons, but damage from these sources may closely follow the accompanying gamma ray dose.
- There is no evidence for a change in scintillation in efficiency due to radiation damage from gammas.
- There is no significant dose rate effect for gammas. However, there may be a dose rate effect for hadrons.
- *There is a strong, radiation induced* phosphorescence with an emission at ~ 340 nm.
- There is a strong thermoluminescence signal with an emission at about the same wavelength as the phosphorescence.
- *There is very slow, but measurable, recovery at room temperature for* irradiated crystals kept in the dark.
- Damage from gammas, neutrons and hadrons are all thermally annealable. Thermal annealing in dry nitrogen produces no undesirable side effects, but annealing in air causes diffusion of oxygen and hydrogen into the the crystal through the absorption of moisture.
- Nearly full recovery can be achieved by illumination to visible and/or UV light. The amount of light required for optical annealing is relatively small.

4. Analytical Techniques and Material Analysis

A number of very powerful analytical techniques were employed to study the the impurities and defects in crystals which could give rise to radiation damage. Some of the principle methods used were SIMS (Secondary Ion Mass Spectroscopy), GDMS (Glow Discharge Mass Spectroscopy) RBS (Rutherford Backscattering), EPR (Electron Paramagnetic Resonance) and TL (Thermoluminescence). Each of these techniques provides a different type of information which together can be used help understand the impurity and defect structure of the crystals.

Figure 6 shows an image produced using SIMS [8] which shows the location of an inclusion of hydrogen and oxygen in a crystal produced at SIC. The bright areas at top of each image shows the correlation of both of these elements in the same region, indicating the likely presence of OH^- in the crystal. The presence of oxygen and hydrogen was also confirmed using EPR in a number of other samples [9].

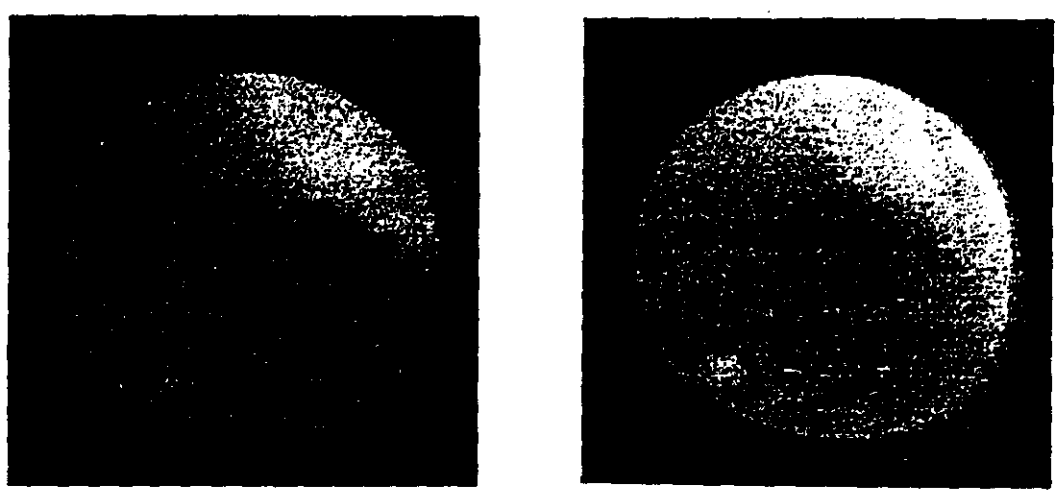

Fig.6 *Secondary Ion Mass Spectroscopy (SIMS) image of a 50 μm diameter region of a BaF_2 crystal [8]. Bright areas at the top of both images are inclusions of hydrogen(left) and oxygen(right).*

A detailed trace elements analysis was performed on many samples, and it was found that certain elements were present in surprisingly large concentrations. In particular, in

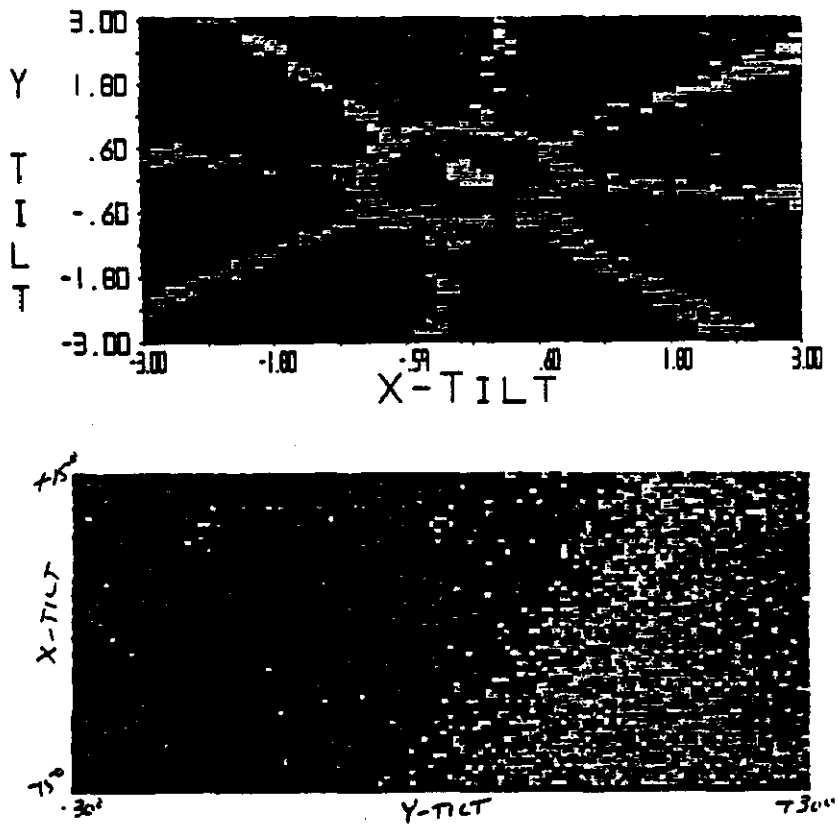

Fig.7 *Rutherford Backscattering (RBS) image for good (top) and poor (bottom) quality crystal surface [8]. Line pattern in the top figure is due to ion channeling in a highly ordered crystal structure.*

some of the earlier crystals produced at SIC, strontium was found to present at a level greater than 1%. Other elements, such as chlorine and sulphur, we present at the tens or hundreds of ppm level. Rare earth and transition metal impurities were generally at the level of a few ppm or less, but could be more harmful as far as radiation damage, even in smaller concentrations. The concentration of many of these impurities was greatly reduced by using higher purity raw materials and by improving the vacuum in the growth furnaces. However, it is particularly difficult to remove oxygen from any fluoride crystal due its similarity between fluorine and oxygen in the lattice. Therefore oxygen, either in molecular form, or in the form of OH^-, plays an important role in the radiation hardness of BaF_2, as discussed in the next section.

The importance of the surface quality was also studied along with its relationship to radiation hardness [10].Figure 7 shows RBS images of two crystals with different qualities of surface polish. The upper image is of a good quality surface which shows a set of

crossing lines produced by ion channeling through a highly ordered crystal structure. The lower image is of a poor quality surface, which shows no ion channeling, and indicates an amorphous layer at the surface. However, although the quality of the crystal surface may have other important implications for the overall performance of the detector, no definite correlation was found between poorer quality surface polish and radiation damage.

5. Principle Causes of Damage

Although the investigation carried out by the GEM collaboration was quite extensive, and a tremendous amount of information was obtained on the effects of radiation damage in BaF_2, no definite conclusions can yet be drawn on the exact cause of damage in the material. It is, however, generally accepted that BaF_2 is intrinsically very radiation hard, and that the effects seen are due to impurities or defects in crystal. A great deal has been done to identify certain impurities which are believed to most likely be the cause of specific types of damage. For example, certain rare earths such as Ce, Pr, Tm and others, are know to cause absorption bands at particular wavelengths after irradiation [11-12].

However, certain key elements such as oxygen and hydrogen, seem to play a particularly important role in radiation damage. The effect of oxygen appears to be an increase in the induced absorbance in the 200-220 nm region after irradiation. This is precisely the most detrimental region region in terms of its affect on the scintillation light output. A model for the mechanism for this damage has been proposed by a group at West Virginia University [9], and independently by a group at Tonji University in China [13].In this model, OH^- ions are present in the crystal due to the absorption of H_2O during growth and processing. Exposure to radiation dissociates the OH^- and creates O^- ions along with interstitial hydrogen atoms (H_i^o). At the same time, F-centers are created independently in the crystal during irradiation. After a certain time, some of the interstitial hydrogen atoms move to the F-centers and recombine to give substitutional hydrogen ions (H_s^-). From calculations done by the Tongji group, the H_s^- ions are responsible for an absorption band at 206 nm, and the O^- ions for another band at 240 nm. These calculations clearly need further confirmation, but they do indicate a likely problem with hydrogen and oxygen in terms of radiation damage.

6. Conclusions

It is clear that improving the purity and reducing the number of defects would tend to reduce the amount of radiation damage produced in BaF_2, as it would in any crystal. The level of some impurities, such as rare earths and transition metals, can be reduced by higher purification of the raw material, but at the expense of increasing the cost of the finished crystal. Others, such as oxygen and hydrogen, which are introduced during processing, are much harder to eliminate. These can be reduced by improved growth and processing techniques, such as using cleaner furnaces, higher vacuum, etc. Defects can also play a major role in the sensitivity to radiation damage, which are largely affected by processing, such as growth conditions and annealing. However, it is virtually impossible to completely eliminate all impurities and defects in any crystal, and it therefore becomes a question of how sensitive a particular material is to these various inhomogeneities which will ultimately determine its radiation hardness. Given this fact, it is clear that finding a practical solution for dealing with a certain level of radiation damage in particular a application is extremely important.

Because of its critical nature, a panel of experts in the area of radiation damage in various materials was convened by the SSC lab to review the situation of radiation damage in BaF_2 for the GEM experiment. The panel consisted of 14 members from the areas of solid state physics, materials science, high power laser optics, and crystal growth. The charge of the panel was "to review the current level of understanding of radiation damage in BaF_2 crystals and the progress in producing appropriately radiation hard crystals in China". The panel met three times and reported their findings to the SSC laboratory and to the GEM collaboration [14].Their basic conclusions were that they found no obvious fundamental reason why $BaF2$ could not be made sufficiently radiation hard to meet the GEM requirements, and that the primary issue regarding radiation hardness was that of purity and quality. However, at the time of its last meeting in August 1992, crystals of adequate radiation hardness had not yet been produced. In view of that, the panel concluded that the only solution for dealing with the problem of radiation damage in the GEM experiment was that of optical annealing in situ. This solution was intensively studied by

GEM, and in September, a subgroup of the Expert Panel concluded that "optical annealing of radiation damage in BaF_2 with low intensity, visible light can essentially eliminate the radiation induced absorption of the fast component of the scintillation light" [15].It further concluded that the current quality of the production crystals available in China could meet all of the GEM specifications. However, for reasons which are beyond the scope of this report, the GEM experiment adopted an alternative technology for its electromagnetic calorimeter. Nevertheless, this study of radiation damage in BaF_2 has been a prime example of how a detailed, systematic study can lead to a vastly improved understanding of radiation effects, and will hopefully be continued for BaF_2 as well as other materials.

7. References

1. H.Newman, contribution to these proceedings.
2. *A Report on Radiation Damage in BaF_2 to the GEM Collaboration*, and references therein, C.L.Woody, GEM TN-91-38, Dec., 1991.
3. C.L.Woody et.al., IEEE Trans. Nucl. Sci. NS-39 (1992) 123.
4. *On Quality Requirements to the Barium Fluoride Crystals*, R.Y.Zhu, GEM TN-92-48, Jan. 1992.
5. *Radiation Damage in Large Barium Fluoride Crystals*, P.J.Li et.al., GEM TN-92-77, March 1992.
6. *On the Optical Bleaching of Barium Fluoride Crystals*, D.A.Ma and R.Y.Zhu, GEM TN-92-149, Sept. 1992.
7. *GEM Detector BaF_2 Precision Calorimeter Selection Report*, GEM TN-92-170, Aug. 1992.
8. Images and analysis produced by M.Strathman of Charles Evans and Associates, Redwood City, Ca..
9. *Identification and Characterization of Radiation Induced Point Defects in BaF_2*, L.E.Halliburton et.al., West Virginia University Technical Report No. 3, July, 1992.
10. *Barium Fluoride Research and Development at Lawrence Livermore National Laboratory*, C.R.Wuest, B.A.Fuchs and W.Kway, GEM TN-93-163, July 1992..

11. *Radiation Damage of Rare Earth Ions Doped Barium Fluoride Crystals*, G.Chen et.al., GEM TN-92-78, March 1992.

12. C.L.Woody et.al.,IEEE Trans. Nucl. Sci. NS-36 (1989) 536.

13. *Radiation Effects in Hyrdrolyzed Barium Fluoride Crystals*, L.Y.Chen et.al., GEM TN-92-129, June, 1992.

14. Reports of the GEM BaF2 Panel: Dec. 4-5, 1991, Jan. 21-22, 1992 and Aug. 3-4 1992.

15. Report of the BaF_2 Expert Group, GEM TN-92-173, Sept. 1, 1992.